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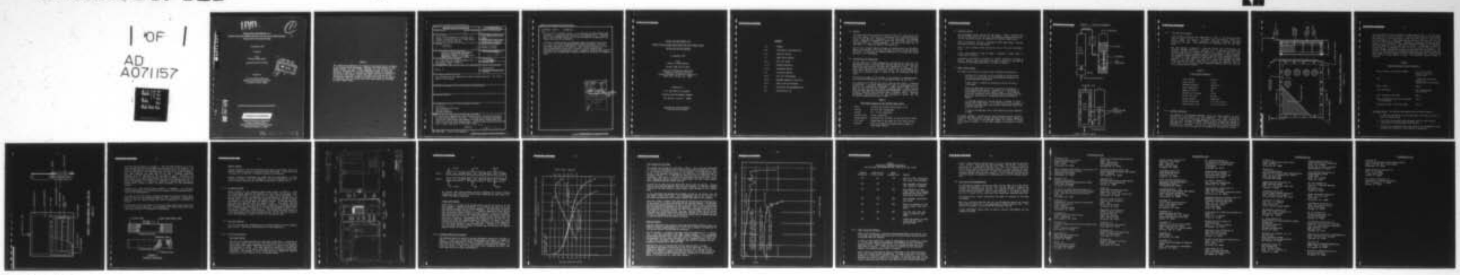

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DESIGN AND DEVELOPMENT OF A 30 WATT SOLID POLYMER ELECTROLYTE FUEL CELL POWER SOURCE FUELED WITH CALCIUM HYDRIDE

12 December 1978

O. Adlhart

Final
Technical Program Report

Contract DAAK 70-77-C-0222

Prepared for

U. S. Army Mobility Equipment
Research and Development Command
Fort Belvoir, Virginia 22060

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ENGELHARD

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Calcium Hydride
Hydrogen Generator

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

In recent years MERDCOM has shown renewed interest in the Solid Polymer Electrolyte Hydrogen-Air Fuel Cell. It focuses on the broad spectrum of applications where power requirements are quite small ranging from fractions of a watt to a few hundred watts. Handling simplicity and versatility are of utmost importance in this power range.

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ABSTRACT CONT'D (BLOCK 20)

The SPE cell is uniquely suited. It is operative at about ambient temperatures, is rugged and has excellent life characteristics. Combined with a solid hydrogen source such as calcium hydride or magnesium high energy densities are attainable.

A 24 Volt, 30 Watt device was developed under the contract consisting of a static SPE stack integrated with a hydrogen generator utilizing cartridge contained calcium hydride. Even in short missions of a few hours energy densities are well in excess of those obtained with secondary batteries. Additional work is needed to optimize the system for operation over a broad range of ambient conditions including low temperatures.

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CONTENTS

1.0 Summary

The report describes design and evaluation data for a hydrogen-air SPE fuel cell system. The device is integrated with a hydrogen generator using calcium hydride - contained in a fuel cartridge - as hydrogen source. Hydrogen is generated by direct but restricted contact of the fuel with water in a "Kipp" generator. Its flow to the fuel cell is regulated in a "dead end" mode of cell operation.

The 28 volt, 30 watt device provides an energy density of 60 WH/kg. Based on the weight of the fuel cartridge in excess of 1000 WH/kg are delivered. If the water for hydride conversion is included, the energy density is still 550 WH/kg.

2.0 Introduction and Objectives

Contract DAAK 70-77-C-0222 represents a continuation of a SPE fuel cell development effort initiated under contract DAAK 53-76-C-0130 with the design and fabrication of 30 three watt hydrogen-air cells. The program proved that bipolar construction features of the phosphoric acid stack can be applied to the SPE stack design. The current program provides for a scale-up from a 3 watt, 5 volt to a 30 watt, 28 volt stack.

An additional aspect of the program is the design of a hydrogen generator using calcium hydride and its integration with the fuel cell.

The combination of the SPE cell with a hydride fuel offers -- compared to batteries -- increased energy density, ease of rechargeability and superior low temperature performance. Under contract DAAK 70-77-C-0222, two 28 volt, 30 watt devices integrated with a calcium hydride based hydrogen generator were delivered to MERADCOM. Two 24 volt units without a generator were supplied under a contract option. Required performance characteristics for the hydrogen-air stacks and the integrated system are outlined in Table 1.

Table 1

Performance Objectives for Hydride Power Source

Power	30 watts at 40-110°F and 20 watts at 0°F
Voltage	24 to 32 VDC, unregulated
Current	0.1 - 1.25 A adjustable
Start-up time	15 min. above 40°F
Energy density	550 WH/kg fuel and water at any load above 3 watts
Tilt angle	Operational at any angle up to 45° from vertical
Mission	4 hrs at 30 watts and proportionally longer at lower power levels

3.0 Technical Effort

The development effort consists of two phases. Phase 1 involves the design and fabrication of fuel cell and generator subsystems and their testing separately as well as combined to a complete power source.

Based on evaluation results, a redesign follows under Phase 2 and two deliverable devices are fabricated.

Phase 1 and 2 designs differ materially only in the stack configuration.

A dual stack system is used in Phase 1, whereas a single stack is relied upon on Phase 2.

Information discussed in Section 3.1 refers primarily to Phase 2 design. However, evaluation data discussed in Section 3.2 are derived from testing performed under Phase 1 as well as Phase 2.

3.1 Power Source Design

The power source design is based on the following considerations:

> Reactant air to the fuel cell is supplied by diffusion and air access is restricted to prevent membrane dehydration at elevated ambient temperatures.
>
> Product water is removed by evaporation and/or drained as liquid water.
>
> Waste heat generated in the fuel cell and by reaction of calcium hydride with water -- combined, it is equivalent to approximately two times the electric ouput -- is dissipated by natural convection and at elevated ambient temperatures by additional cooling with a thermostatically controlled air blower.
>
> For hydrogen generation, calcium hydride is brought in direct contact with liquid water. It is contained for this purpose in a tubular cartridge with provision for liquid water access and hydroxide expansion.
>
> To control the hydrogen flow a self-regulating "Kipp" generator is used.

A systems schematic indicating the above mentioned design features, including the mode reactant supply and waste heat removal appears in Figure 1. The filter in the hydrogen supply line serves to absorb trace amounts of ammonia. It was later removed as discussed in section 3.2.2.

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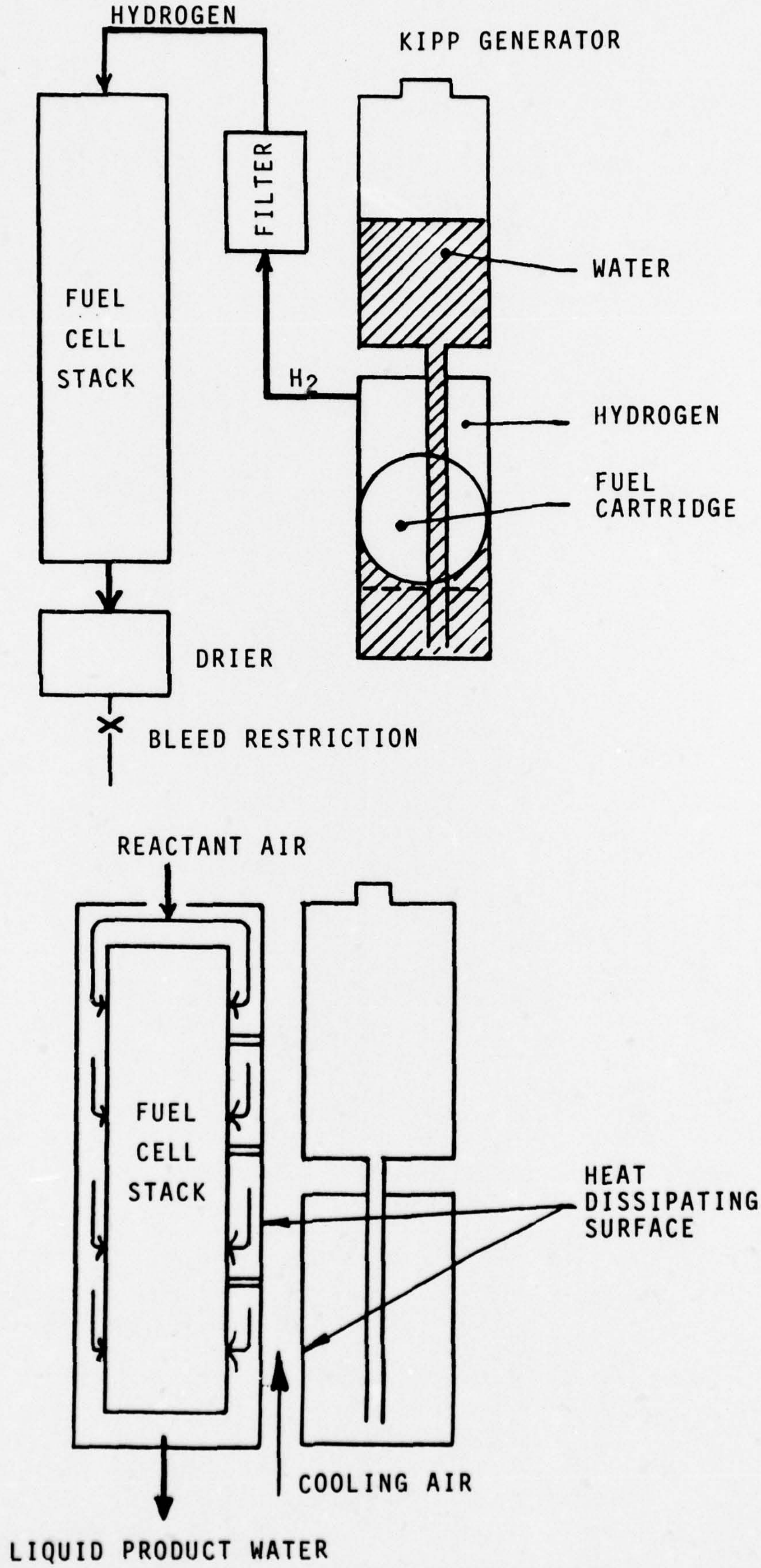
FIGURE 1 - SYSTEMS SCHEMATIC



3.1.1 Fuel Cell Stack Design

Stack design parameters are summarized in Table 2 and detailed design features appear in Figure 2. A bipolar cell construction, carbon collector plates and carbon substrate supported, platinum activated electrodes are used. The electrodes are laminated with the SPE membrane.

The stack design is based on a long and narrow cell configuration and external hydrogen manifolds. Reactant air is supplied by diffusion. Hydrogen, supplied at a pressure of 5-20 cm W.C., enters one manifold at the stack top and exists the opposite manifold at the bottom. Stack components are assembled between aluminum channels rigidized with honeycomb inserts. The assembly is held together by perforated face plates attached to these channels. The same plates are used also to press the hydrogen manifolds firmly against the stack. Waste heat is rejected by convection from the stack and a heat dissipating plate connected thermally with the stack by five extended bipolar plates.

Table 2

Stack Design Parameters

Stack voltage	24-32 V
Stack current	1.25 A
Hydrogen pressure	$\leq$25 cm W.C.
Stack temperature	$\leq$125°F
Number of cells	34
Nominal cell voltage	0.70 V
Active cell area	48 cm^2
Current density	26.$mA.cm^{-2}$
Bipolar plate size	3.1 x 23 x 0.4 cm
Cooling plate size	3.7 x 23 x 0.4 cm

3.1.2 Hydrogen Generator

The design of the hydrogen generator relies on direct contact of solid hydride with liquid water in a "Kipp" generator. This mode of operation assures a high rate of hydrogen generation even from a small quantity of fuel and quick response to demand changes without intermediate storage of hydrogen gas. In order to control the reaction with water the hydride is contained in a cartridge. The use of a cartridge also simplifies handling of the moisture sensitive material and disposal of the hydroxide residue.

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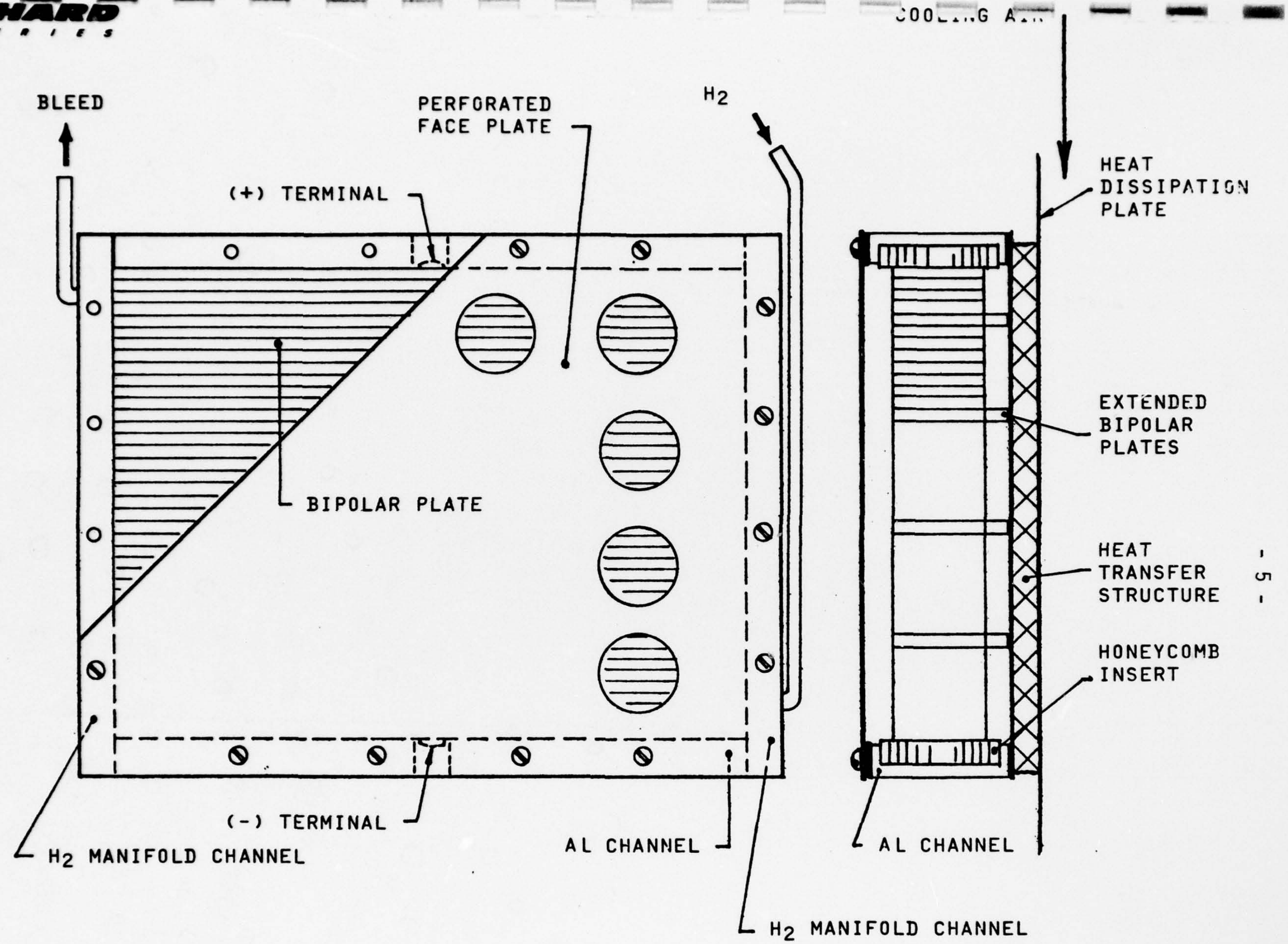


FIGURE 2 - FUEL CELL STACK, FRONT & SIDE VIEW

(SCALE 1/2)

Design parameters of the generator are summarized in Table 3. The principle components are the water reservoir, reaction chamber and fuel cartridges. The generator is self regulating. Water from the reservoir flows to the reaction chamber via a tube connecting the bottom of the reservoir with the bottom of the reaction chamber. Hydrogen is generated as soon as water reaches the fuel cartridge. The rate of generation depends on the degree of cartridge immersion and is controlled as follows: If hydrogen is generated faster than it is consumed in the fuel cell, pressure increases in the reaction chamber forcing water partly or fully back into the reservoir; conversely, if more hydrogen is consumed than generated, the pressure drops and water flows from the reservoir into the reaction chamber thus immersing the cartridge to a larger degree in water. The generator design is shown in Figure 3.

Table 3

Hydrogen Generator Design Parameters

Water reservoir and reactor chamber	12.5 x 28 x 3.8 cm aluminum container, 1330 ml each
Fuel	-4 mesh calcium hydride >93% purity contains up to 1% nitrogen
Water charge	900 ml
Fuel cartridges	Two aluminum tubes, 3.8 x 25.5 cm
Fuel charge per cartridge	140g
Water consumption upon full conversion (2 cartridges)	470 ml
Expansion volume in each cartridge	120 ml

Cartridge Design: The hydride cartridge has the following features:

1. A container consisting of a thin wall metal tube with a series of holes for water entry.
2. Lining of the container with absorbent material such as glass fiber paper for uniform water distribution.
3. Provision for expansion within the container to accommodate volume changes upon conversion of the hydride to hydroxide.

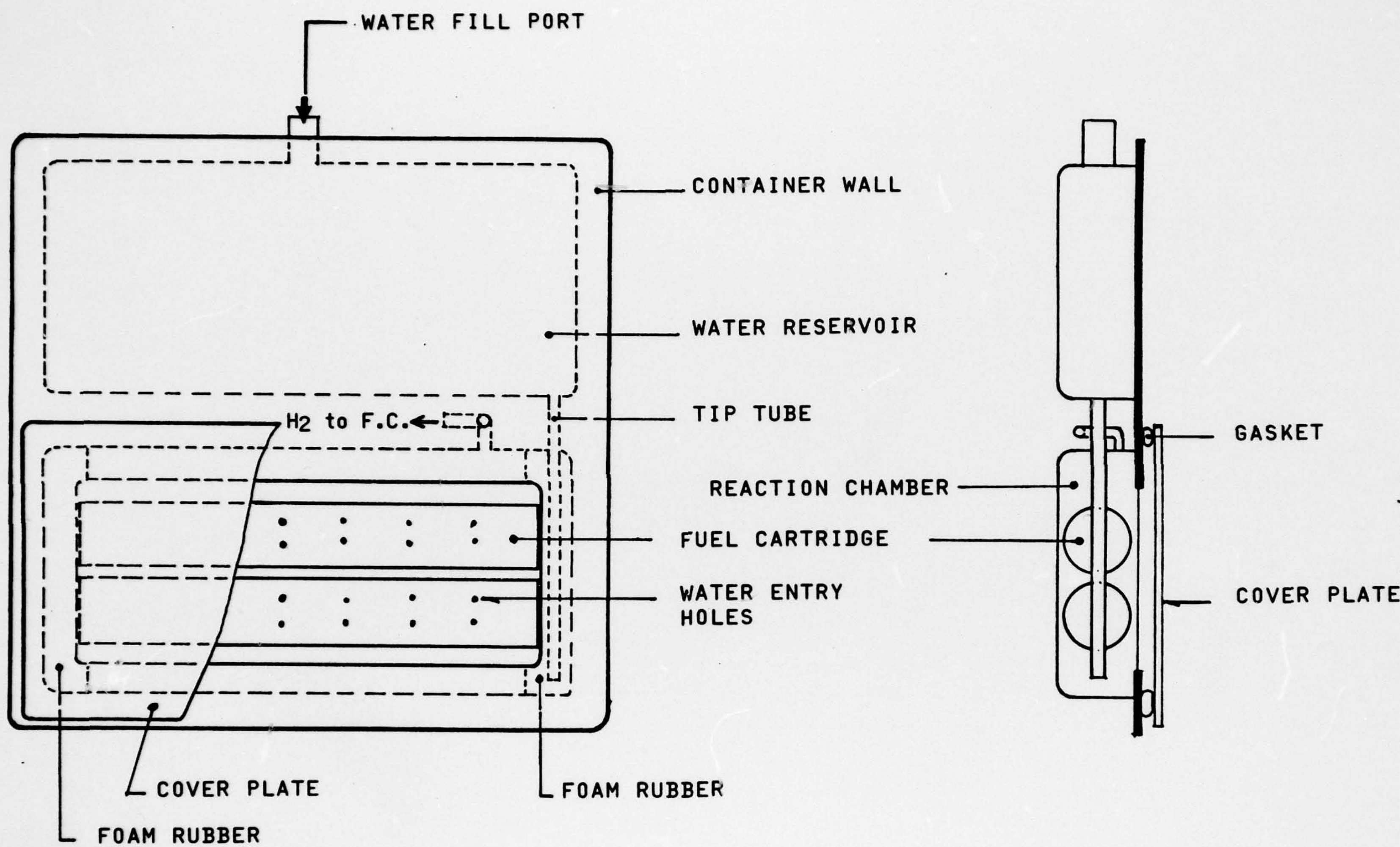


FIGURE 3 - HYDROGEN GENERATOR, FRONT & SIDE VIEW

(SCALE 1/3)

Cartridge design appears in Figure 4. The cartridge consists of a 25 cm long aluminum tube of 3.8 cm diameter with 20 holes of 0.7 mm diameter. The tube is lined with glass fiber paper and filled to 2/3 of its volume with -4 mesh calcium hydride. The remaining space is taken up by plugs of polystyrene foam inserted from both ends of the tube. Hydrogen generation commences with the immersion of the cartridge in water. Initially, the water enters at a rate controlled by the size and number of holes and is distributed by the glass paper lining contacting the hydride bed over its entire circumference. As hydride is converted, a porous hydroxide cake is formed which provides a path for water transport to the unconverted hydride.

Formation of a cake with suitable porosity is important. It very much affects completeness of conversion and the degree to which hydrogen generation rate can be regulated.

If porosity is too low, water transport through the cake may become inadequate to substain the required generation rate; conversely, if hydroxide particles are too loosely packed, the rate of generation is difficult to control.

The provision for volume expansion in the cartridge assures the formation of a cake with suitable porosity. It accommodates the volume changes that take place upon conversion of the hydride to hydroxide.

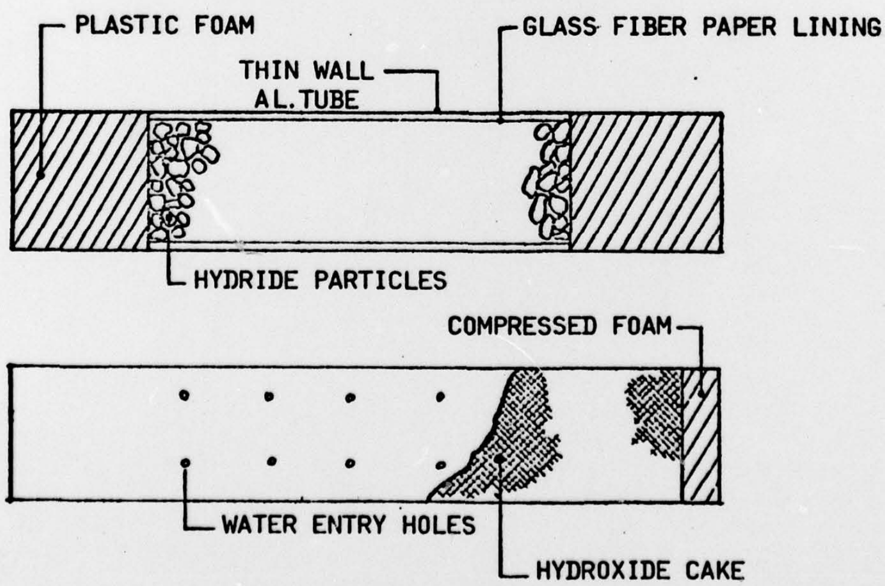


FIGURE 4

HYDRIDE CARTRIDGE

Ammonia Removal

Hydrogen generated from calcium hydride may contain appreciable amounts of ammonia formed by reaction of calcium nitride with water. (Calcium nitride is present as an impurity in technical grade calcium hydride).

Removal of ammonia is necessary for stable long term performance. A filter containing sulfonic acid resin (Amberlyst 15) was considered for this purpose but found unsuitable and modified as described in section 3.2.2.

3.1.3. Integrated System

Design features of the integrated power source appear in Figure 5. Fuel cell and hydrogen generator are mounted on 1.5 mm thick, 30 cm square aluminum sheets which make up the wall of the power source container. Air blower, ammonia filter and bleed gas drier are mounted on the same plate as the fuel cell. Filter and drier are accessible from the outside to permit replacement of the drying agent and ammonia scrubber. A flexible hose connects the Kipp generator with the ammonia filter. From there, purified hydrogen enters the cell stack at the top. Bleed gas is released from the stack bottom to the bleed gas drier and from there through a porous metal disc to the atmosphere. Reactant air is supplied through nine holes of 1.6 cm diameter in the top of the container. Air reaches the cells through a .75 cm gap between cell stack and container wall on one side and an 0.6 cm thick perforated heat transfer structure on the opposite side.

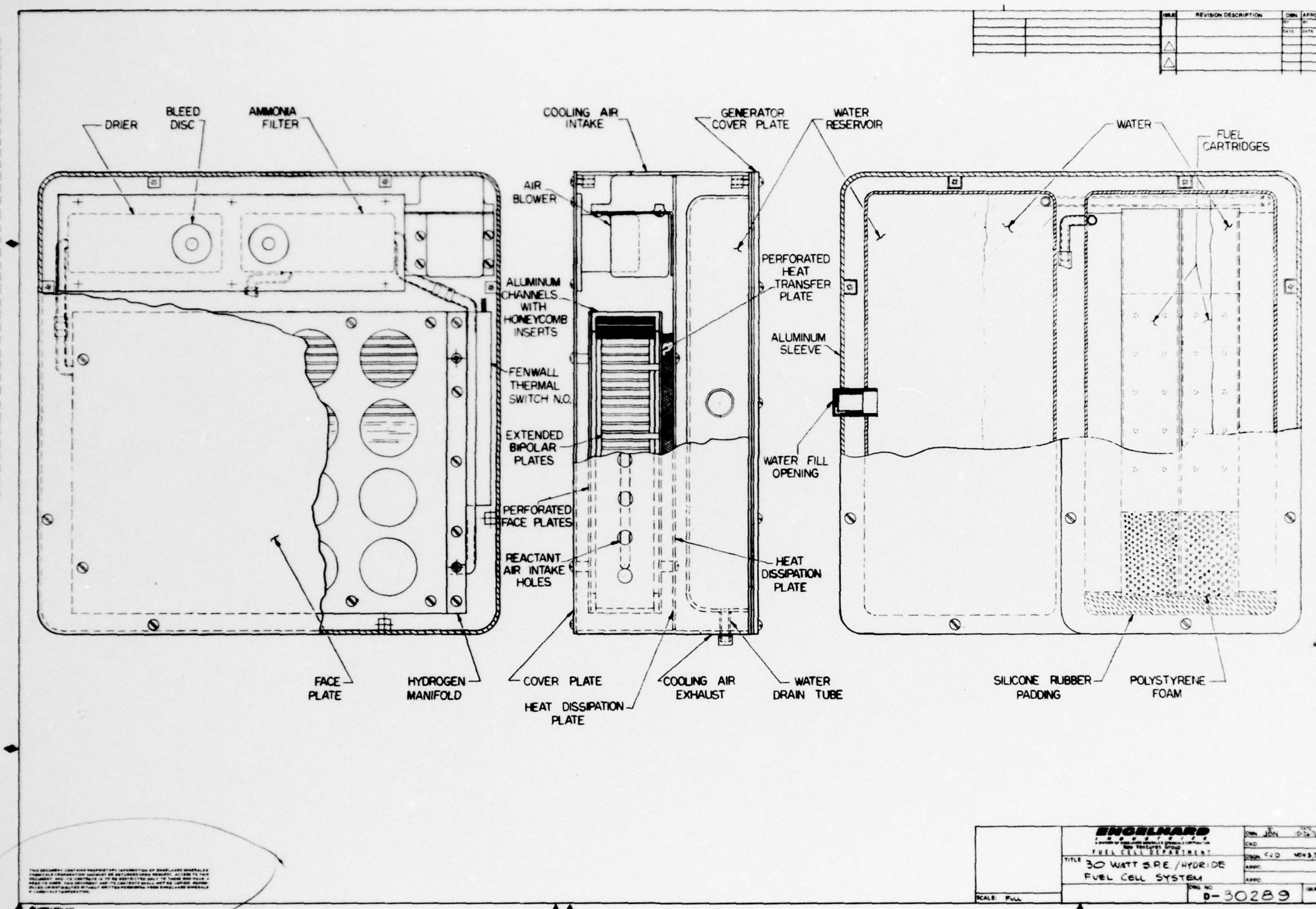
3.2 Evaluation Results

Fuel cell and generator subsystem were evaluated separately and as complete power source. Testing was performed only at room temperature.

3.2.1 Fuel Cell Evaluation

Dual Stack Design

The fuel cell system designed and fabricated under Phase 1 consisted of two 34 cell stacks with an active cell area 20 cm^2 (9.1 x 2.2 cm) each. These stacks were tested connected electrically in parallel and supplied with hydrogen in series. This mode operation resulted in a significant performance decline and uneven load sharing due to thermal and moisture imbalances. The changes in stack voltage and stack temperature observed during a 20 hr test (load 20 Ω) are illustrated below: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FENWALL THERMAL SWITCH N.O.
EXTENDED BIPOLAR PLATES
PERFORATED FACE PLATES
REACTANT AIR INTAKE HOLES
PERFORATED HEAT TRANSFER PLATE
ALUMINUM SLEEVE
WATER FILL OPENING
HEAT DISSIPATION PLATE
FACE PLATE
HYDROGEN MANIFOLD
COVER PLATE
HEAT DISSIPATION PLATE
COOLING AIR EXHAUST
WATER DRAIN TUBE
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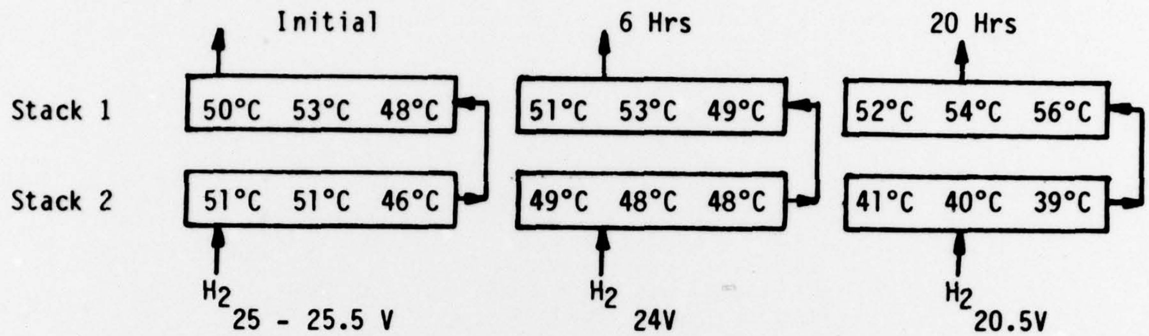


For uniform load sharing among parallel connected SPE stacks, thermal integration and limitation of the current supplied by each stack to a common load appears necessary.

Single Stack Design

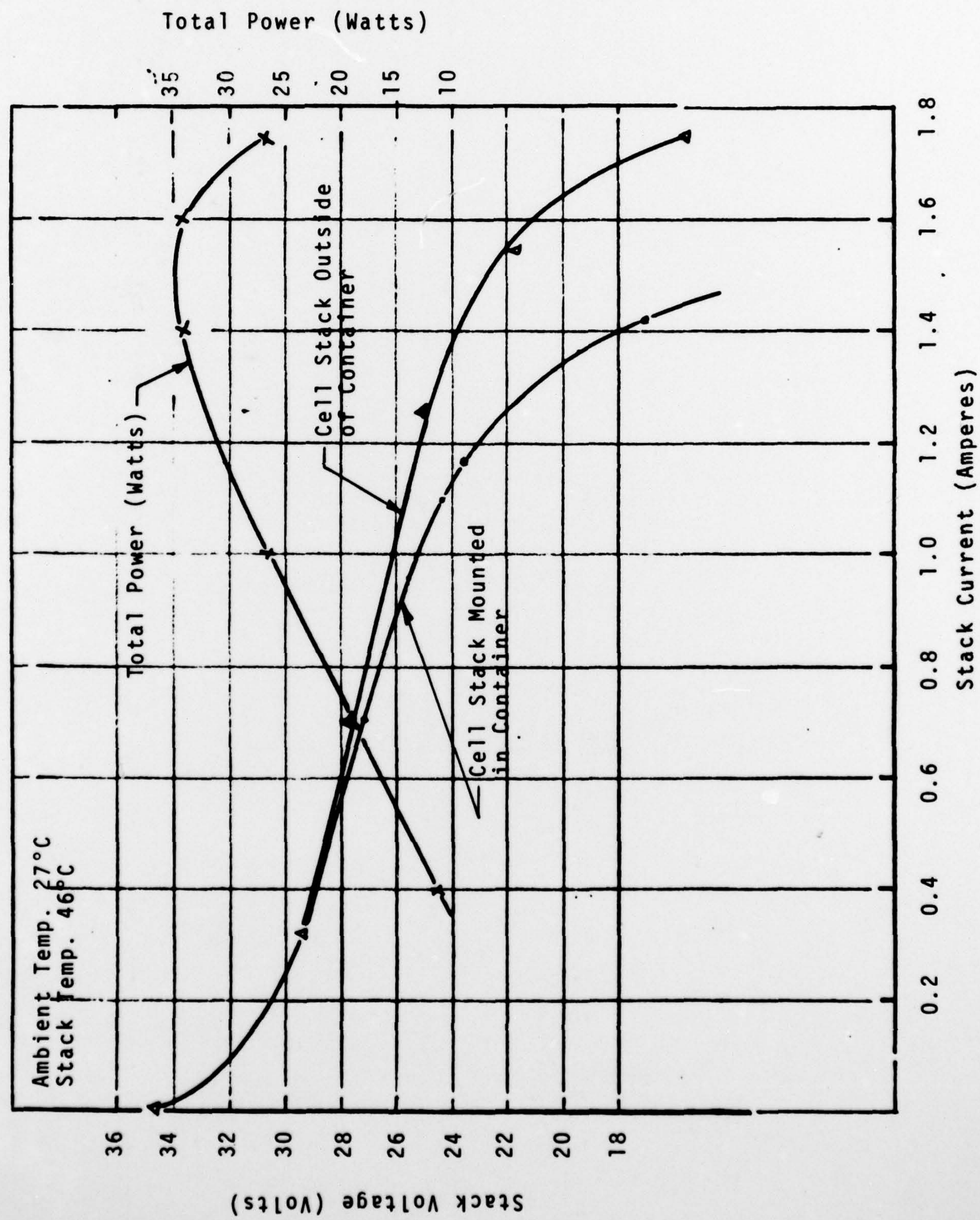
Under phase 2, a single stack design with an active cell area of 48 cm^2 was developed. Temperature gradients in this design are small due to the high thermal conductivity of stack components. At rated load, variation over the entire stack are below 3°C. Gradients between the stack and the heat dissipating surface (Figure 2) range from 5-7°C. Most of the temperature drop takes place in the contact area between the bipolar plate and the face plate and is due to a coating applied to the bipolar plate for electrical insulation. Performance data representative for the four (4) stacks built under this program appear in Figure 6. At rated load, stack voltage varies by less than 1 volt. When the stacks are fully exposed to air, the output maximum is less than 30 watts. Changes in the design of the face plate and heat transfer structure may be required to improve oxygen transfer to the stack.

3.2.2 Hydrogen Generator Evaluation

The hydrogen generator meets or exceeds performance objectives outlined in Table 1. Start up is essentially instantaneous once water is added and hydrogen can be generated in excess of maximum load requirements up to a tilt angle of 45° from vertical. The hydride is utilized fully; however, in order to sustain a load of 30 watts and fully discharge the fuel, two cartridges have to be used simultaneously.

FIGURE 6 - CURRENT-VOLTAGE CHARACTERISTICS OF SPE POWER SOURCE

(See Table 2)



Fuel Charge Per Cartridge

The optimum fuel charge per cartridge is 140 g. This value was determined in a series of experiments which are discussed below and summarized in Table 4. As indicated in Section 3.1.2, full utilization of the hydride and regulation of the hydrogen flow depend on the formation of a hydroxide cake of suitable porosity. It is obtained by providing room for expansion in the cartridge. This space is filled with polystyrene foam which, while gradually compressed during conversion, always keeps the fuel firmly packed, thus, preventing excessive water take up.

Location of the foam does not materially effect fuel utilization. Similar results were obtained whether the foam was inserted in intervals between portions of the fuel charge or simply used to plug the ends of the aluminum tube.

In one experiment the hydride was arranged in form of an annular ring and the center filled with foam. This arrangement may be of interest for cartridge configurations with larger diameter than used in this program.

Test data listed in Table 4 were obtained by discharging one cartridge only. It was placed 2.5 cm above the bottom of the reaction chamber. The hydrogen generated was metered directly or fed to the fuel which in these experiments was not thermally integrated with the generator. The results obtained from the discharge of one cartridge differ from the simultaneous discharge of two cartridges in the degree of water take up. Two cartridges take up proportionately more water because the cartridge located at the bottom of the reactor is discharged first, but continues to absorb water although hydrogen is supplied mostly from the second cartridge.

Ammonia Content

Hydrogen generated from technical grade calcium hydride (Ventron Corp.) was found to contain approximately 1.3×10^{-3} val % ammonia corresponding to 0,25 mg per gram of calcium hydride converted.

For its removal, a scrubber cartridge containing Amberlyst 15, a sulfonic acid resin, was provided in the power source design. This arrangement proved unsatisfactory since under most conditions hydrogen is released from the generator not only water saturated but at a temperature well above that of the filter cartridge. Water condenses from the moist hydrogen causing plugging or by-passing of the resin bed.

Admixing the resin to the hydride directly does provide a suitable alternative and permits elimination of the filter cartridge. Thus, by adding 20 g Amberlyst 15 to a cartridge charge of 140 g -4 mesh calcium hydride, the amount of ammonia released was reduced to 0,05 mg/g from 0,25 mg/g. This may be further reduced by better dispersion of the absorbent which is utilized only to a very small degree.

FIGURE 7 - DISCHARGE CHARACTERISTIC OF HYDRIDE POWER SOURCE

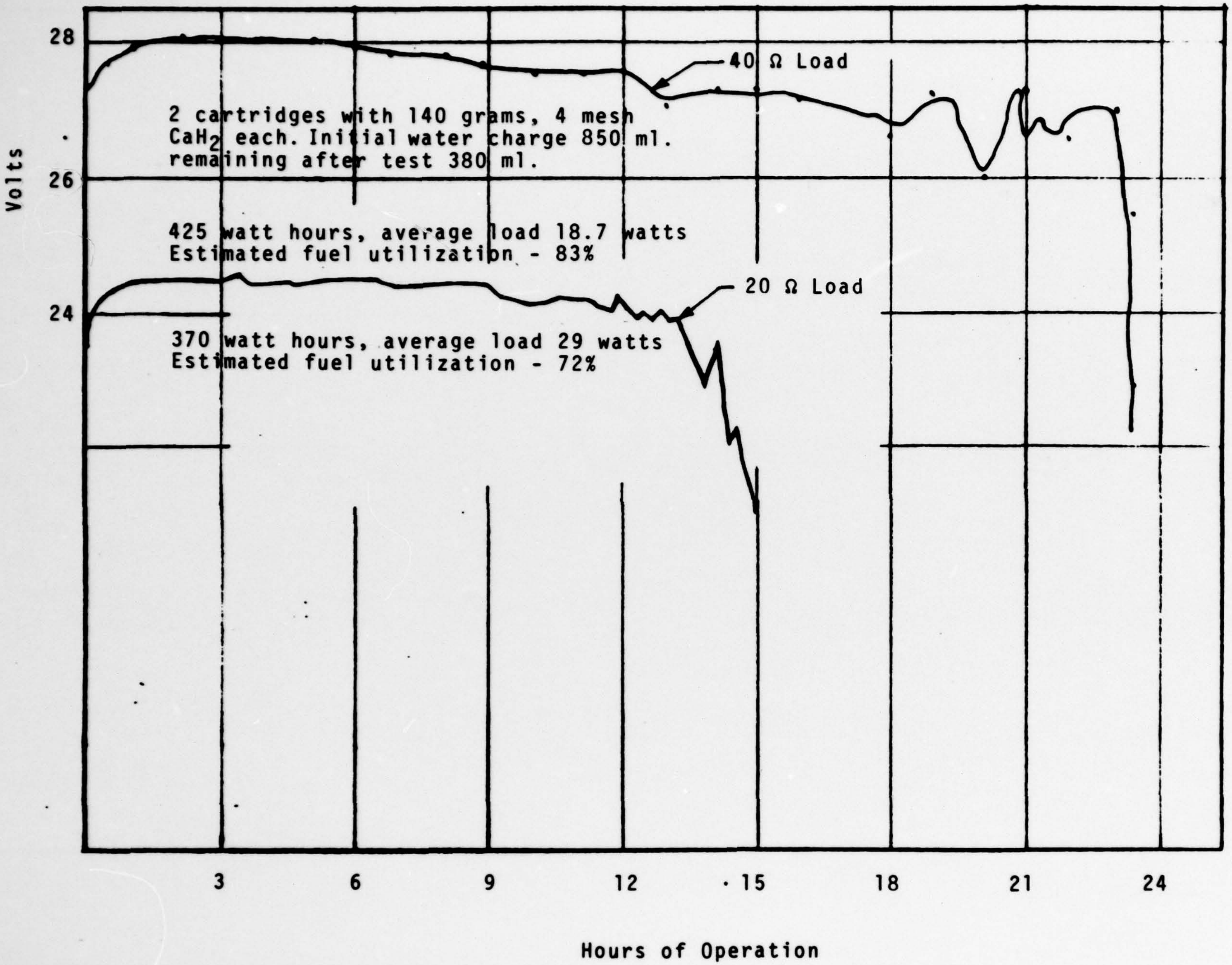


Table 4
Hydride Utilization Experiments
One Cartridge, Discharge with 40 Ω Load to 20 Volt Cutoff

Hydride Charge (g)	Watt Hrs to 20 Volt Cutoff	Water Consumed (ml)	Remarks
120	121	170	Fuel cell leak, polystyrene, foam plugs, fuel consumed. ,
130	173	--	Fuel consumed, polystyrene foam inserted in segments.
130	150	310	Poor regulation, 9 additional leaks, fuel consumed, hydroxide cake water soaked polystyrene foam plugs.
140	194	200	Fuel consumed, polystyrene foam plugs.
140	187	230	Annular arrangement of fuel foam in center, fuel used - fully.
140	108	220	Fuel cell leak, fuel consumed, polystyrene foam plugs.
160	133	160	Significant amount of uncon verted hydride, dense hydroxide cake.

3.2.3 Power Source Performance

Power source evaluation, including discharge experiments over periods of up to one day, was performed with only one of the two 30 watt hydride devices fabricated under the program.

In tests at room temperature, capacity requirements are exceeded by a factor of 3-4 and energy density requirements of 550 WH/kg for fuel acid water combined are met. Power output at 24 volts is below 30 watts due to the restricted air supply. If the fuel cell is removed from the container however, the required output level is reached.

Quick start-up is observed and power is delivered in less than one minute rather than fifteen minutes permissible. A higher purge rate is initially necessary at least with a new fuel cartridge. During start-up, fuel cell an generator are quickly purged but air trapped in a new fuel cartridge is only slowly released and does lead to inert gas build up at normal purge rates.

Figure 7 shows discharge characteristics of the integrated hydride system fo a 20 Ω and 40 Ω load using two fuel cartridges. At the 20 load level, additional air is supplied to the fuel cell. This test, therefore, does not reflect acutal power source performance, however, it does indicate the capacity of generator and fuel cell to meet the required performance level. Discharge voltage varies within a range of 2 volts and some fluctuation was observed towards the completion of discharge due to erratic hydrogen generation.

4.0 Conclusion and Recommendations

The development program carried out under contract DAAK 70-77-C-022 demonstrated the suitability of the SPE cell for the design of compact and reliable air breathing power sources. Its combination with a fuel such as calcium hydride provides -- even in short missions -- an energy to weight ratio far in excess of that available from conventional secondary batteries.

The system offers further convenience and speed of recharge by cartridge replacement.

Waste heat available from the fuel cell and the hydrogen generating reaction should permit operation at low temperatures without capacity loss. This possibility remains to be confirmed by additional testing.

Further development should focus on specific mission requirements and low temperature design.

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